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Critical review on uranium and arsenic content and their chemical mobilization in groundwater: A case study of the Malwa region Punjab, India

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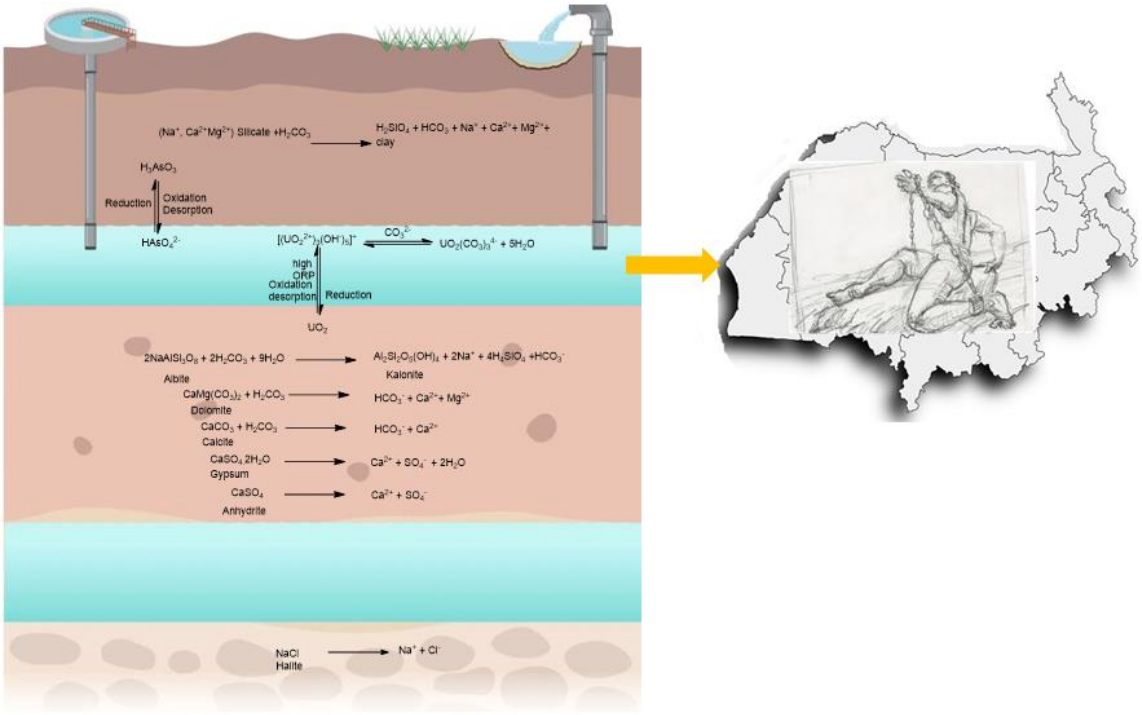
Abstract

The presence of pollutants like uranium and arsenic in the groundwater can have a terrible impact on people's health (both radiologically and toxicologically) and their economic conditions. Their infiltration into groundwater can occur through geochemical reactions, natural mineral deposits, mining and ore processing. Governments and scientists are working to address these issues, and significant progress has been achieved, but it's challenging to deal with and mitigate without adequately understanding the different chemical processes and the mobilization mechanism of these hazardous chemicals. Most of the articles and reviews have focused on the particular form of contaminants and specific sources of pollution, such as fertilizers. However, no literature report exists explaining why particular forms appear and the possible basis of their chemical origins. Hence, in this review, we tried to answer the various

questions by devising a hypothetical model and chemical schematic flowcharts for the chemical mobilization of arsenic and uranium in groundwater. An effort has been made to explain how chemical seepage and excessive groundwater use resulted in the change in aquifers' chemistry, as evidenced by their physicochemical parameters and heavy metal analysis. Many technological advancements have taken place to mitigate these issues. Still, in low-middle-income countries, especially in the Malwa region of Punjab, also known as Punjab's cancer belt, paying a high amount for installing and maintaining these technologies is an unviable option. In addition to working to improve people's access to sanitary facilities and clean water to drink, the policy-level intervention would focus on increasing community awareness and continued research on developing better and more economical technologies. Our designed model/chemical flowcharts will help policymakers and researchers better understand the problems and alleviate their effects. Moreover, these models can be utilised in other parts of the globe where similar questions exist. This article emphasises the value of understanding the intricate issue of groundwater management through a multidisciplinary and interdepartmental approach.

Keywords: groundwater, uranium, arsenic, hydrogeochemistry, chemical mobilization.



Introduction

Water covers three-quarters of the earth's surface, but only 0.3% of water, especially from lakes, rivers, and groundwater, is available for human utilization (Davis and De Weist, 1966; Shiklomanov, 1993). The demand for drinking/agricultural water keeps increasing, so the burden on freshwater resources from great rivers to underground aquifers also increases (Lall et al., 2020). Although these aquifers are renewable, the rate of pumping out water is faster than the rate of recovery; hence, water depletion occurs faster than enrichment (Fendorf and Benner, 2016; MacDonald et al., 2016). Not only quantity but water quality also becomes a major concerning factor. According to the world water report by United Nations, the increasing groundwater withdrawal decreases its quality worldwide. From the total world groundwater content, Asian countries draw a significant share of about 65%; moreover, Indian groundwater resources are highly exploited (UNESCO World Water Assessment Programme, 2022). In the Punjab state of India, where the green revolution occurred, most of the groundwater resources

are overexploited (See supplementary information and (Punjab Water Regulation and Development authority, 2020). Indeed, Jurgens and coworkers demonstrated that overexploitation caused hydrogeochemistry changes and deteriorated the water quality (Jurgens et al., 2010). The leaching from natural chemical deposits and agrochemicals (fertilizers, pesticides, insecticides and herbicides) also contribute to the deterioration of water quality. As a result, surface and groundwater sources are polluted with several toxic heavy metals (Kaur et al., 2019).

One region of Punjab, the Malwa region, was once known as the breadbasket of India, now called the cancer belt of Punjab. The state government agency conducted a study in collaboration with the Indian Council of Medical Research (ICMR) and showed results in this favour (Nanda et al., 2016a). However, people also suffer from various other diseases like Arthritis, Sinusitis, Anemia, Fluorosis, Arsenicosis, Lead poisoning, Methaenoglobinemia and kidney-related problems. Uranium and Arsenic are the two major contaminants present in groundwater, putting a shattering effect on the socioeconomic model of society. Several reports highlighted the presence of these toxic metals in higher concentrations than the WHO recommended limit. But the research's finding describing the flow of these hazardous chemicals inside the earth's surface of the Malwa region is very sparse. Here, we collected more than 100 scientific research papers describing the excess uranium and arsenic content in the groundwater of the Malwa region of Punjab.

Numerous articles and reviews have been published regarding the form of uranium [$UO_2(CO_3)_3^{4-}$ form and $(UO_2)_3(OH)_5^{+}$] found in the groundwater and the cause of contaminations, particularly geogenic/fertilizers. However, none of them included information about the origins and relationships between the origins, for instance, why this form? How was this form formed? What are other possible sources of origin of one particular form? Without the proper knowledge of the mechanism of the whole process, it is difficult for policymakers

to devise the proper control measure. So, intending to help the policymaker, we designed this study as shown in Figure 1.

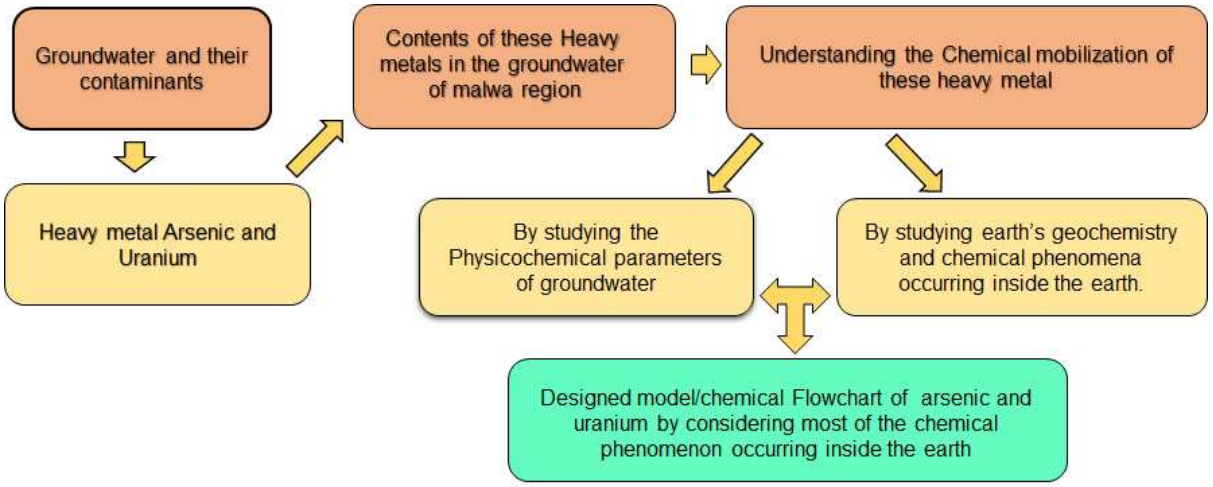


Figure 1. Conceptual framework model of our work

The correlation studies of various physicochemical parameters told that the change in one physicochemical parameter could be the reason for a change in other parameters, but there are very minimal reports describing the process of how this change comes out. So, the main objective of this study is to understand the complex interplay of natural dynamics of the hydrological cycle through the chemical phenomenon materialising inside the earth. Hence, the chemistry treatment to previous studies was given in order to build the connecting links and comprehend the proper mechanism of their flow and proposed hypothetical models for earth geochemistry and chemical schematic flowcharts for the movement of uranium and arsenic inside the groundwater. Here, we limited our study to uranium and arsenic only, but we will try to explore more about other heavy metals, such as cadmium, lead, strontium etc., in our further communications. The schematic flowcharts will explain all possible ways to mobilize arsenic and uranium in the groundwater. This review attempts to underscore the complex interaction of heavy metal (U and As) contaminants in groundwater and help to understand their transport process. In our review, we tried to glimpse the available technologies for removing uranium and arsenic in the groundwater with their merits, demerits and operating

costs. Yadav et al. found in their study that these technologies were unaffordable for low-middle-income countries (Yadav et al., 2022). It's even difficult to manage these technologies for the people of the Malwa region (also known as the cancer belt), where family income is overburdened due to cancer. Here, the average family income is Rs 2.30 lakh compared to the average cost of cancer treatment (Rs 2.75 lakh) (I. Singh et al., 2013). The policy-level intervention would prioritise raising community awareness and ongoing research on creating better and more affordable solutions, in addition to focusing on improving people's access to sanitary facilities and clean water to drink.

Further, the chemical scenario provided by our designed models will be helpful for academicians, scientists and policymakers to understand the problems better and take the necessary steps to mitigate these issues. This study offers the national and state governments strategic guidance on comprehending and addressing the problem of poor water quality in Punjab's Malwa region. Moreover, this paper will highlight the urgent need for interdisciplinary and interdepartmental initiatives to ensure sustainable groundwater quality. Furthermore, the model employed here could be utilized in other areas of the world where groundwater has been seriously polluted.

The Malwa region of Punjab

Punjab is the land of five rivers, but the Sutlej and Beas are the two major rivers that pass through the state, while the Ravi river touches the northern part of the state. This river pattern divides the state into three regions: Malwa, Majha and Doaba. Among the three areas, the Malwa region is the most significant part of the present Punjab state of India. Sutlej river's left bank separated the Malwa region from the other regions. Its southern border is shared with Haryana and Rajasthan, while the western edge is shared with Pakistan. The Malwa region geographically extends from 29° 30' North to 31°10' North and longitudes 73° 50' to 76° 50' east, as shown in Figure 2. It occupies 65.1% (32808 km^2) of the total Punjab area and bears

58% of the Punjab population. Cotton, rice and wheat are the major crops grown in this area, and the region is known as the cotton belt of India (Kaur and Kaur, 2016).

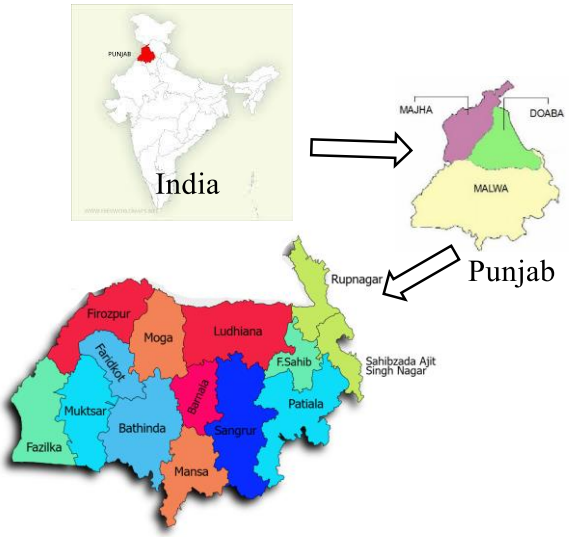


Figure 2. Map of Malwa region of Punjab.

The effect of the green revolution

After the green revolution, the Malwa region of Punjab led from the front to feed the nation and made a discernible change in the economy of the state and the whole country (Khush, 2001 and reference cited there in). This boom in agriculture made farmers of Punjab self-reliant. But, on the other hand, considerable investments in the agriculture sector, an increase in farm mechanization and the excessive use of pesticides for an increase in production changed the green revolution to the greed revolution (Nanda et al., 2016b; Planning Commission of Punjab, 2005). In 2020, the pesticides/insecticides consumption was around 5000 metric tonnes compared to 154 metric tonnes in 1954 (Neel Kamal, 2020). The fertilizer consumption increased to 26.5 million in 2009-10 from 78000 tons in 1965-66 (Sharma and Thuker, 2011). Karam Prakash et al. reported that the fertilizers consumption was 253.94 kg/hectare (Karam Prakash, 2022). The utilization of large amounts of fertilizers/pesticides/insecticides and overexploitation of groundwater (Please see the supplementary information) resources led to

changes in the groundwater cycle and groundwater chemistry of the area, as shown in Figure 3.

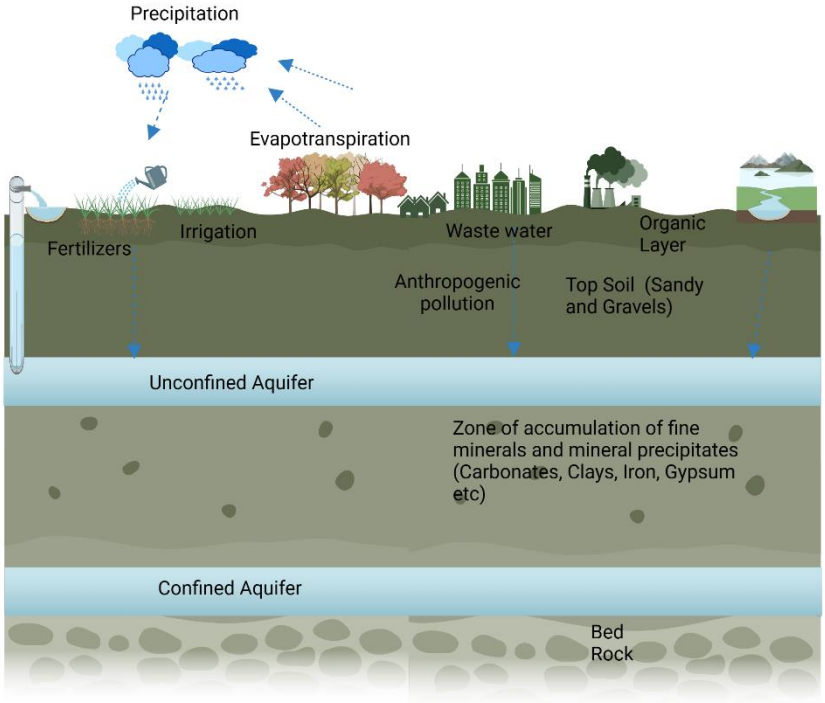


Figure 3. Pictorial representation of the groundwater cycle and groundwater layers of this region

In 1995, Singh et al. reported uranium in the water of Bathinda and Amritsar (Singh et al., 1995). In the late 90s, Philipose et al. published an article in the Indian Express newspaper that described the scenario of Punjab and warned about the future consequences (Philipose, 1998). In 1999, by documenting cancer fatalities and referring to them as "cancer-stricken villages," Pandhar's editorial in the newspaper brought two villages, Gyana and Jajjal of district Bathinda, to national attention (Panher, 1999). In parliamentary question 2003, the government of Punjab (Proceeding of Punjab Vidhan Sabha, 2003) asked the Punjab Pollution Control Board (PPCB) and Post-graduate Institute of Medical Education and Research (PGIMER) Chandigarh to conduct a study in Talwandi Sabo and Chamkaur Sahib blocks of Bathinda and Rupnagar districts, respectively. This study found that females were more affected by different types of cancers than males. Moreover, they reported heavy metals (U, As, and other heavy metals)

contamination in drinking water/groundwater at a higher concentration than the World Health Organization (WHO) permissible limit.

Later on, Thakur et al. displayed the presence of pesticide residue in the vegetables, milk and blood samples of different patients (Thakur, 2005; Thakur et al., 2008). Halder et al., in a survey, reported premature greying of hair, premature ageing and excessive cancer death in the Jajjal village of Bathinda (Haldar, 2007). After these eye-opening reports, many other research groups and organizations worked in this region and published their findings of uranium and arsenic contents in the groundwater, summarized in Table 1.

Table 1. Collection of reports about uranium and other heavy metals content in groundwater of Malwa region

Area of Study	**Metals content found**	**Concentration range (ppb or µg/l)** (1 ppb = 1 µg/l)	**Sample Taken**	**Reference**
Bathinda	Uranium	11.7 - 113 ppb	Groundwater	(Singh et al., 1995)
Bathinda	Uranium	2 - 87.5 µg/l	Groundwater	(Kumar et al., 2006)
Bathinda, Mansa	Uranium	7 - 316 ppb	Groundwater	(Kochhar et al., 2007) (Kochhar et al., 2012)
Malwa Region	Uranium	5.41 – 43.39 µg/l	Groundwater	(Mehra et al., 2007)
Bathinda, Mansa, Faridkot, Firozpur, Sangrur, Moga Patiala	Arsenic	3.5 – 688 µg/l	Groundwater	(Hundal et al., 2009)(Hundal et al., 2007)
Bathinda, Rupnagar	Arsenic	> 10 ppb	Groundwater	(Thakur et al., 2008)
Bathinda, Mansa	Uranium	0.9-63.1 ppb	Groundwater	(Singh et al., 2009)
Bathinda, Mansa	Uranium	28.57 – 213.36 mBq/l	Milk	(Kumar et al., 2009)
Malwa Region	Uranium	>100 ppb	Groundwater	(Muhanad et al., 2009)
Bathinda, Mansa, Faridkot, Firozpur	Uranium	0.2 – 644 µg/l	Groundwater	(Kumar et al., 2011) (Kumar et al., 2014)
Bathinda, Mansa, Firozpur, Faridkot	Uranium	3.2 – 60. 5 ppb	Groundwater	(Prabhu et al., 2012)
Muktsar	Uranium	4.5 – 330 µg/l	Groundwater	(Shenoy et al., 2012)
Bathinda, Faridkot, Firozpur, Sangrur, Muktsar	Arsenic	5 – 50 µg/l, 10 – 100 µg/l, 10 – 50 µg/l, 5 – 50 µg/l, 5 -50 µg/l	Groundwater	(Sharma et al., 2013)
Bathinda	Arsenic	>10 µg/l in 1/3 samples	Groundwater	(Singh et al., 2013)
Bathinda	Uranium	0.48 – 571.7 µg/l	Groundwater	(Singh, L. et al., 2013)
Malwa Region	Uranium	13.9 – 172.8 µg/l	Groundwater	(Tripathi et al., 2013)

Bathinda, Moga, Faridkot	Arsenic	16 – 76 µg/l	Groundwater	(Sidhu et al., 2014)
Bathinda, Mansa, Faridkot, Firozpur, Sangrur, Moga Patiala	Arsenic	2.2 – 120 µg/l	Groundwater	(Shah et al., 2015)
Faridkot, Bathinda, Mansa	Uranium	0.13 – 676 µg/l	Groundwater	(Saini et al., 2016) (Saini et al., 2017)
Bathinda	Arsenic	2.28 – 27.47 µg/l	Soil	(Kumar et al., 2016)
Mansa	Uranium	0.13 – 1340 µg/l	Groundwater	(Sharma and Singh, 2016)
Bathinda, Mansa, Faridkot and Firozpur	Uranium Arsenic	0.5 – 571.7 µg/l 1 – 59.6 µg/l	Groundwater	(Bajwa et al., 2017)
Bathinda, Mansa, Muktsar Faridkot	Arsenic	4.35 – 23.94 µg/l	Groundwater	(Kaur et al., 2017)
Bathinda, Mansa, Muktsar Faridkot Firozpur, Sangrur, Moga, Barnala	Arsenic	2 – 1200 µg/l	Groundwater	(Sharma and Dutta, 2017)(Sharma, 2018)
Mansa, Bathinda	Uranium	2.3 – 357 µg/l	Groundwater	Sharma, DA et al., 2017)(Sharma et al., 2020)
Faridkot, Muktsar	Uranium,	3 – 190 µg/l	Groundwater	(Pant et al., 2017)(Pant et al., 2020b)
Bathinda, Mansa, Firozpur, Faridkot	Uranium	1.78 – 261 µg/l	Groundwater	(Singh, KP et al., 2018a)
Ludhiana	Arsenic	0 – 21 µg/l	Groundwater	(Singh et al., 2019)
Ferozpur, Patiala, Rupnagar	Arsenic	16 – 91 µg/l	Groundwater	(Virk, 2019a)
Bathinda, Mansa, Faridkot, Firozpur, Sangrur, Moga Patiala	Uranium	2.47 – 366 µg/l	Groundwater	(Virk, 2020; Virk, 2019b, 2019c, 2019d, 2019e, 2018, 2017a, 2017b, 2017c)
Bathinda	Arsenic Uranium	2.1 – 83.87 µg/l 8.98 – 289.53 µg/l	Groundwater	(Kaur, G et al., 2021)
Bathinda, Barnala, Ludhiana	Arsenic Uranium	0.5 – 28.7 µg/l 0.5 – 432 µg/l	Groundwater	(Kumar et al., 2021)

Table 1 shows that high uranium content was found in districts such as Bathinda, Mansa, Faridkot, Ferozpur and Moga district. Furthermore, from the data provided by CGWB 2021, the spatial and temporal distribution map and a 3D bar of uranium graph were made, as shown in Figure 4. These graphs provided a vivid illustration of the seriousness of the situation.

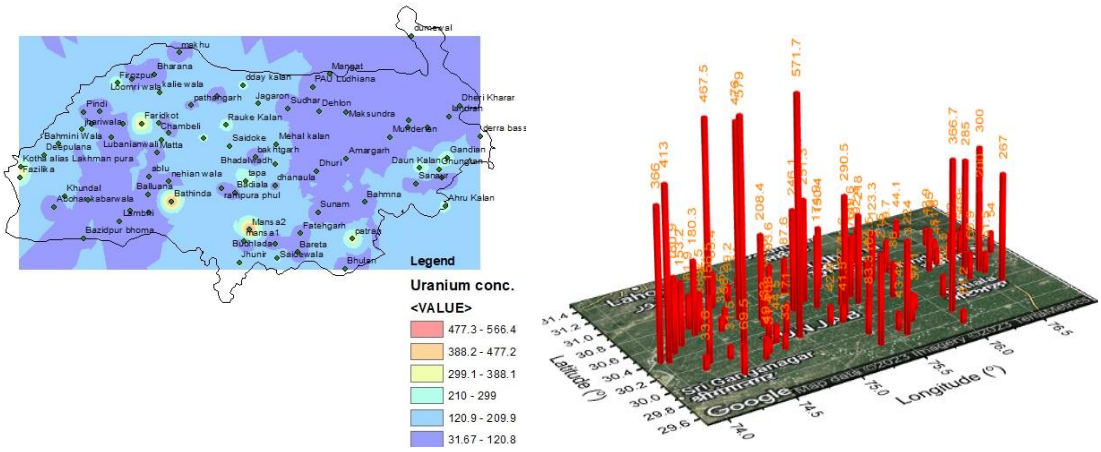


Figure 4. Spatial and temporal variation and 3D bar graph of uranium distribution in the Malwa region of Punjab.

This issue of high uranium contamination in the groundwater came to the limelight after Prof. Carin Smit, a South Africa-based clinical toxicologist, visited Baba Farid Centre (BFC) for special children, where children are being treated for autism, cerebral palsy and neurological impairments. He took samples and analyzed them. He reported high Uranium, Barium, Cadmium, Manganese, and Lead contents in the patients' samples (Blaurock-Busch et al., 2010; Blaurock-Busch et al., 2010). This finding became a front-page headline in various national and international newspapers, for instance, The Times of India, Down to Earth, The Telegraph (London) etc. Hence, Center Ground Water Board (CGWB), Punjab Pollution Control Board (PPCB), Punjab Water Supply Sanitation Department (PWSSD), Punjab State Planning Board and other government and non-government agencies were employed to learn more about the situation. In the study by Punjab State Planning Board, Punjab, India, Bhabha Atomic Research Center, India (BARC) and Guru Nanak Dev University, Punjab, India (GNDU), the researchers found the excessive use of phosphate fertilizers as a possible source of the high amount of uranium in the region by percolation through the soil (Bajwa et al., 2017; Kumar et al., 2011). Still, Srivastava et al. and Singh et al. reports ruled out that hypothesis (G. Singh et al., 2018; Srivastava et al., 2017). After that, other theories were put forward, such as fly ash dump of the thermal power station, industrial effluents, etc. However, Alrakabi et al. 2012 suggested that

the most plausible origin of high uranium content in the Malwa region might be geogenic (Alrakabi et al., 2012). This high uranium content in the groundwater can cause severe kidney problems, lung infections, autoimmune disorders, high blood pressure, reproductive system problems, and cancer (Domingo, 1994; Leggett, 1994; Leggett and Pellmar, 2003).

According to the Ministry of Agriculture, India report-2013, 6500 metric tonnes (MT) of pesticides were consumed only in Punjab, and 75% was used up in the Malwa region of Punjab alone (Ministry of Agriculture, 2014). Researchers published many reports about pesticide contents in drinking water, food and vegetables, and all the studies are compiled in Mittal's review article (Mittal et al., 2014). Arsenic is a primary component of many pesticides and is typically present in the form of lead arsenate, sodium arsenate, calcium arsenate, dimethyl arsenate, chromated copper arsenate, fluorochrome arsenate phenoyl, etc. (Bencko and Yan Li Foong, 2017). However, Hundal et al. found that in the Malwa region of Punjab, hand pumps and canal water are also more heavily contaminated with arsenic than is permitted by the WHO and put light upon the geogenic origin of arsenic in groundwater. (Hundal et al., 2009, 2007). They reported that the arsenic contents in Patiala, Bathinda, Muktsar, Mansa, Faridkot, and Firozpur districts exceeded the permissible limit. A study by PGIMER and PPCB reported that the Buddha nullah river became highly toxic due to effluents from industries and contained various harmful heavy metal ion content such as As (Machhan, 2019). From table 1 and other literature data, the Mansa district of the Malwa region was found to have very high arsenic contents with an average value of 650 µg/l (Sharma et al., 2021b). Interestingly, some other locations in the southwestern part of Malwa found a relatively high arsenic content. Hence, we found an interesting trend in the arsenic ion concentration in the Malwa region. Considering the average value of arsenic in different districts of the Malwa region, a spatial distribution trend map and 3D bar graph of arsenic ion concentration in groundwater were plotted for understanding, as shown in Figure 5.

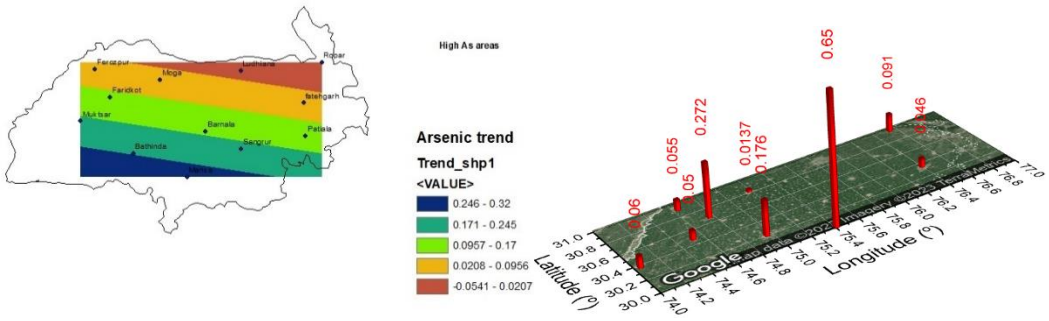


Figure 5. Spatial and temporal variation and 3D bar graph of arsenic distribution in the Malwa region of Punjab.

Arsenic is a deadly poison considered a carcinogenic element that causes cancer through respiratory and gastrointestinal exposure. The study conducted by Hong and co-workers confirmed the association between lung, skin and bladder cancer due to arsenic poisoning (Hong et al., 2014). Recent studies linked arsenic with other types of cancer, such as liver, prostate, leukaemia, etc. (Martinez et al., 2011). The relationship between arsenic and various diseases, such as diabetes, neurological effects, cardiac disorders, congenital disabilities and reproductive organs, was also found (Claudia et al., 2003; Lee et al., 2002; Tsai et al., 2003; Tseng et al., 2002). Consequently, it is imperative to study the mechanism of mobilization of these dangerous metals from top to bottom.

Physicochemical parameters of groundwater of Malwa region

To understand the mobilization of arsenic and uranium, we first studied various physicochemical parameters and correlations. The information about the quality of water and its suitability for drinking use are easily obtained by analysing the physicochemical parameters, such as pH, electrical conductivity (EC), total dissolved solutes (TDS) and turbidity and by calculating the inorganic and organic components in the water and influence of biotic and abiotic factor (Kumar et al., 2007). The inorganic elements are essential for various body functions, but their higher concentrations create groundwater pollution and human health-related issues. Various natural and anthropogenic processes such as the leaching of chemicals,

rocks weathering, mining, chemical fertilizers and the metal industry can be the reason for deviation of these parameters (Thakur et al., 2016). Moreover, these parameters mentioned above vary with weather changes, such as the areas' premonsoon and post-monsoon (CGWB, 2020, 2019, 2018). These variations might be due to the difference in the geological location of the study area and the method used for sample collection. So here, the physicochemical parameters data of groundwater from recent publications/reports about all districts of the Malwa region are compiled as shown in Table 2.

The pH data from Bathinda (6.9 -9.5), Mansa (7.5–9.1), Faridkot (7.0–9.8), Muktsar (6.8-9.0) and Fazilka (7.8 -9.6) districts showed slightly inclination toward the upper limit of WHO permissible value (Table 2). The pH value is affected by the presence of carbon dioxide and various inorganic ions. Electrical conductivity speaks for the measure of the total dissolved ions/salts and salinity. The high electrical conductivity and high TDS data of the southwest part of the Malwa region {Bathinda [Ec(223 -3870), TDS(164-2500)], Mansa [Ec(268 – 5140), TDS(160-3400)], Faridkot [Ec(814 – 7542), TDS(446-4600)], Muktsar [Ec(513 - 11500), TDS(303-5785)] and Fazilka [Ec(745 - 8320), TDS(600-6800)]} is only due to dissolution or leaching of aquifer mineral or mixing of saline source or both respectively (Hounslow, 2018). Due to the very high EC and TDS, the land of the Muktsar and Fazilika districts became infertile. No crop has been sown in the Rattakheda and Sikhwala villages of Muktsar district and Shajrana village of Fazilka district since the last two decades (Shah, 2013). Total hardness depends upon calcium, magnesium, carbonate, sulphate, and chloride ion concentrations. Table 2 showed a high value for TH (0-1490), which may be due to the calcareous texture of the soil. According to the Davis-DeWeist classification for TDS (Davis and De Weist, 1966) and Durfor-Becker classification of TH (Durfor and Becker, 1964) (Table 3), the groundwater of the Malwa region comes in the category of very hard water and unfit for drinking.

Table 2. Physiochemical parameters of groundwater of all the district of the Malwa region of Punjab.*

PP / Districts	pH	Ec	TH	CO_3^{2-}	TDS	HCO_3^-	NO_3^-	SO_4^-	F^-	Ca^{2+}	Mg^{2+}	Na^+	In addition to CGWB, 2021 Other Ref
Bathinda	6.9 - 9.5	223 - 3870	60 - 1125	0 - 156	164 - 2500	85 -818	5.28 -245	0.5 -201	029 -4.79	20 - 285	50 - 900	8.5 - 1120	(Sharma et al., 2021a)
Mansa	7.5 - 9.1	268 - 5140	50 - 1440	0 - 204	160 - 3400	100 -1062	3.6 - 71	5 - 548	0.4 - 2.0	12 - 60	2 - 77	43 -1100	(Sharma et al., 2021b)
Faridkot	7.0 - 9.8	814 - 7542	50 -767	0 - 190	446 - 2700	104 -756	1 - 711	28 -1379	0.23 -4.2	1 - 182	2 - 132	41 -1397	(Ahada and Suthar, 2018)
Firozpur	8.0 - 8.9	478 - 1641	105 - 273	0 - 102	246 - 1740	118 -370	2.4 -172	31 -305	0.14 - 1.21	13 - 46	13 - 51	37 - 310	(Ahada and Suthar, 2018) (G. Kaur et al., 2019)
Muktsar	6.8 - 9.0	513-11500	179 - 761	0 - 263	303-5785	74-816	9 - 2000	31 - 2500	0.39 – 6.4	16 - 408	17.3 - 348	34 - 1123	(Pant et al., 2020a)
Sangrur	7.7 – 9.1	329 - 1715	90 - 560	0 - 156		150 - 683	5.7 - 105	12 - 175	0.15 – 1.2	4 -48	17 - 106	19 - 385	(Ahada and Suthar, 2018)
Patiala	7.6 – 8.9	355-4060	20 - 821	0 - 84		171 - 573	0.35 - 358	5 - 1022	0.19 – 4.12	4 - 72	2 - 156	20 – 650	(Ahada and Suthar, 2018)
Rupnagar	7.1 – 8.5	330 - 1701	57 - 833	0 - 27	199 - 983	70-409	0.14 - 53	37.7 - 522	0.11 – 1.03	13 - 220	3 - 138	15 - 387	(Ahada and Suthar, 2018)
Ludhiana	6.7 – 8.3	80 - 1940	60 - 695	0 - 96	57 - 1370	110 - 696	0.5 - 209	1 - 258	0.08 – 2.75	15 - 250	15 - 620	6.7 - 235	(Kumar et al., 2021)
Moga	7.5 – 8.8	73 - 2332	60 - 495	36 - 132	352 - 1472	134 - 549	107 - 163	20 - 59	0.09 – 10. 5	8 - 98	9 - 102	36 - 260	(Shashi and Bhardwaj, 2011)
Fatehgarh Sahib	6.7 – 8.5	206 - 1452	110 - 650	0 - 27	146 - 1109	168 - 629	0.5 - 65	12 - 120	0.05 – 0.65	15 - 88	30 - 60	30 - 122	(Kumar et al., 2020)
Mohali	7.7 – 9.0	450 - 6480	95 - 1490	0 - 14		182 - 699	0.4 - 407	0.1 - 880	0.31 – 1.52	8 - 360	14 - 163	30 - 820	(Ahada and Suthar, 2018)
Fazilka	7.8 – 9.6	745 - 8320	74 - 911	0 - 240	600-6800	74 - 574	1.5 - 253	43 - 2005	0.32 – 3.1	8 - 206	5 - 418	110 - 1600	(Ahada and Suthar, 2018)
Barnala	6.8 – 8.6	41 - 2340	20 -825	12 - 72	29 - 1657	159 -317	0.5 - 242	1 -392	0.37 – 2.3	10 - 225	10 - 650	30 -182	(Ahada and Suthar, 2018)
WHO recommended Values	6.5 - 8.5	750 - 2000	0 - 500	0 - 500	0 - 1000	-	0 – 50	0 - 400	0.6 – 1.5	0 - 100	0 - 50	200	(Ahada and Suthar, 2018)

*Most recent published data is used to make this table (CGWB, 2020)

Except Ph and Ec(uS/cm), all other parameters are in mg/l

A report by the Planning Commission of India showed the considerable area of four districts of southwestern Punjab, such as Fazilka, Muktsar, Bathinda, and some parts of Mansa, is facing waterlogging and salinization problems, as shown in Figure 6 (Shah, 2013). Moreover, except Rupnagar (0.14 – 53 mg/l) and Fatehgarh sahib (0.5 -65 mg/l) district, a high average nitrate content than the WHO permissible limit in all other districts of the Malwa region was reported (Aulakh and Malhi, 2005), and this is due to excessive use of fertilizers and pesticides, organic and other human wastes (Aulakh et al., 2009).

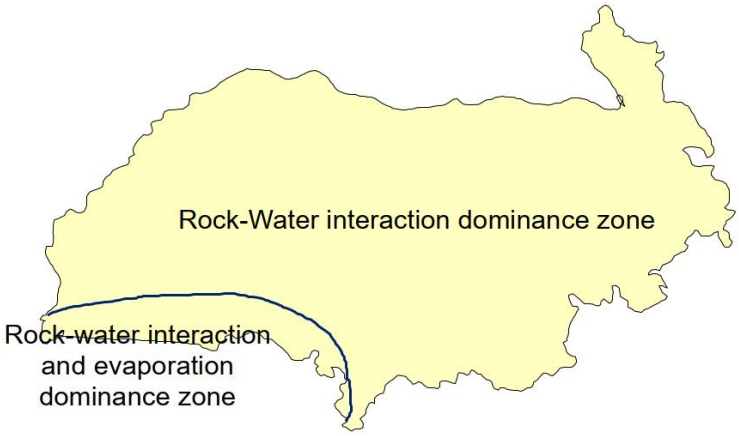


Figure 6. The hypothetical sample model shows the waterlogging area in the Malwa region's southwestern part (Shah, 2013), where evaporation also played a significant role in the change in the aquifer's chemistry in addition to rock-water interaction.

Table 3. Classifications of groundwater based on the physicochemical parameters TH (Durfor and Becker, 1964) and TDS (Davis and De Weist, 1966).

Sr. No	Parameters	Range	Water class
1	TH (Durfor and Becker, 1964)	0 -60 60 - 120 120 - 180 >180	Soft Moderately Hard Hard Very Hard
2	TDS (Davis and De Weist, 1966)	< 500 500 - 1000 1000 - 3000 >3000	Desirable for drinking Permissible for drinking Useful for irrigation Unfit for both

In 9 districts, Bathinda (0.29 -4.79 mg/l), Mansa, Faridkot, Muktsar, Patiala, Ludhiana, Moga, Fazilka and Barnala, the average value of fluoride ions is above WHO permissible limit that may be due to fluoride bearing minerals, fluorite, in aquifers (Wenzel and Blum, 1992). The high bicarbonates and sodium ions concentrations increase the fluoride pollution in groundwater (Kumar and Singh, 2015a). The highest content of fluoride ions (10.5 mg/l) was reported in the Moga district. Lower and higher concentrations of fluoride ions have severe health implications. Its lower concentration below 0.5 mg/l leads to tooth decay, but a higher concentration above 1.5 mg/l causes dental fluorosis (Rathore et al., 2017). The sodium ions, commonly called salinity's indicators, were found in higher concentrations than the recommended limit of WHO (200 mg/l) in most Malwa districts except Fatehgarh Sahib and Barnala (Ahada and Suthar, 2018). The weathering of feldspar minerals and the fertilizers are the most common source of sodium. The high sulphate ions concentration than WHO permissible limit (400 mg/l) in the Faridkot (28 -1379 mg/l), Muktsar (31 – 2500 mg/l), Mansa (5 – 548 mg/l), Patiala (5 – 1022 mg), Fazilka (43 – 2005 mg/l) and Mohali (0.1 – 880 mg/l) districts were reported (Kaur et al., 2017). The breakdown of organic substances from the weathered soils, human activities, fertilizers and pesticides may be the reason for its excess.

The physicochemical parameters study of groundwater quality generated large and perplexing data. Therefore, software-based statistical techniques such as pearson's correlation analysis, principal component analysis, hierarchical correlation analysis, etc., are frequently used to predict the common origin and sources of contaminants in the groundwater (Ofungwu, 2014). These statistical techniques, called multivariate statistical analysis, inform about correlation and variance among the variables and find common factors responsible for pollutants in the water from the complex datasets. Table 4 summarises the linear correlation among different physicochemical parameters and is compiled for understanding. For example, electrical conductivity is attributed to various total dissolved ions (cation and anions), total hardness,

calcium, sodium and chloride concentration. Thakur *et al.* delineated the correlation of high EC with increased dissolved salt content (Thakur et al., 2016), and Tubonimi et al. 2010 described the correlation of total dissolved (TDS) with other ions such as sodium. The positive correlation of sodium ions with Ec and TDS gave information about the salinity content of the soil. The correlation analysis showed that TH is mainly due to calcium and magnesium, along with carbonates, sulphate and chloride. Through these studies, it can be realistic to expect that weathering of limestone, dolomite and other calcium-rich minerals dissolution frequently occurring in the aquifer of this region and the reason for the high carbonate and bicarbonate contents in the groundwater (Tubonimi et al., 2010).

Interestingly, these physicochemical parameters also influenced the uranium and arsenic concentration in the groundwater. Sharma et al. showed a positive correlation between uranium and high TDS in the Mansa district (Sharma et al., 2021b). Through an in-depth study, Sharma et al. found a strong correlation between uranium and total alkalinity. They claimed that high alkalinity might be one of the reasons for the mobilization of uranium in groundwater (Sharma et al., 2019). Hundal and co-workers showed that various geochemical conditions influenced the arsenic concentrations in groundwater (Hundal et al., 2007). The strong correlation between arsenic contamination with a high concentration of iron, phosphate, ammonium ions and anthropogenic activities was demonstrated by Kumar and colleagues (Kumar et al., 2010). Geographically, the Kasoor district of west Punjab of Pakistan lies adjacent to the Malwa region and Afzal and colleagues performed an in-depth study about heavy metals in this district. They revealed the highly erratic distribution of heavy metals in groundwater, which might be brought on by geochemical and anthropogenically-induced polluting sources, such as fly ash from thermal power plants, vehicle pollution, pesticides and fertilisers, corrosion of pipes, chemical industries (Afzal et al., 2014). Inclusively, the concentration of uranium and arsenic ions in groundwater is dependent upon various geochemical processes and conditions such as

oxidation-reduction, associated or competing ions, pH, dissolved salts, alkali content, arid environment and anthropogenic factors etc. Contemporary researchers also use multivariate analysis techniques to find the correlation between different metal ions and their sources of origin. The strong positive correlation among these ions indicates their common origin and source of groundwater contamination in the study area. So, to understand the geochemical changes, there is a need to understand the mechanism of hydrogeochemistry.

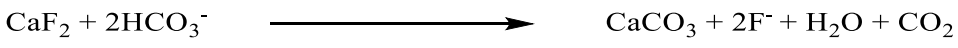
Table 4. Linear correlation analysis among different Physico-chemical parameters.

Sr. No.	Physicochemical parameter	Positive correlation with other parameters	Information derived out	Reference
1	Ec	TDS, Total alkalinity (TA), TH, Ca^{2+}, Cl^{-}, Na^{+}	High EC informs about the mechanism of groundwater circulation, surface infiltration and cation exchange	(Jothivenkatachalam et al., 2010)
2	TDS	EC, TA, TH, Ca^{2+}, Cl^{-}, $Na^{+,}$ K^{+}	TDS delineates some features of precipitation, atmospheric temperature, evapotranspiration and saline intrusion in underground sources	(Tubonimi et al., 2010)
3	TH	TDS, EC, TS, TA, Ca^{2+}, Mg^{2+}, K^{+}	TH tells about the calcareous rock dissolution and ion exchange in the aquifer	(Thakur et al., 2016)
4	Ca^{2+}	EC, TDS, TA, TH	High Ca^{2+}may be due to calcium-rich minerals in the aquifer	(Zare Garizi et al., 2011)
5	Na^{+}	EC, TDS, Cl^{-}	High sodium ion concentration talks about the salinity	(J. Kaur et al., 2021)
6	F^{-}	Na^{+}, pH, HCO_3^{-},	High fluoride and high pH lead to high HCO_3^{-} ion concentration and dissolutions of fluorite.	(Li et al., 2015)

The mechanism controlling the groundwater chemistry: hydro-geochemical evolution

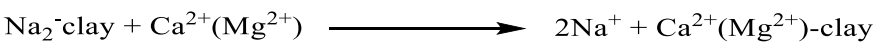
The Malwa region comes under the Indo-Gangetic plains, and its north-eastern part is at a higher elevation than the southwestern region (Jain, 2014). The groundwater flows from higher to lower elevations, so there is a strong probability that the mobilization of minerals from the Himalayas influences the Malwa region's underground water quality (CGWB, 2017). The minerals-water interactions affected the groundwater chemistry. Multiple techniques, for instance, Gibbs plot, saturation index and ion exchange index, etc., are used to study the various hydrogeochemical processes, such as precipitation, rock-water interaction, and evaporation (Feth and Gibbs, 1971; Gibbs, 1970)

To establish the relationship between water composition and aquifer lithological characteristics, Gibbs draws a graph between TDS and cation ratio [$Na^+/(Na^+ + Ca^{2+})$] and anions ratio [$Cl^-/(Cl^- + HCO_3^-)$] and explains the mechanism that controls the groundwater chemistry. Through the Gibbs plot's study, the various research groups (Kaur et al., 2017; Kumar et al., 2021; Thakur et al., 2016) suggested that in the Malwa region, the groundwater chemistry of shallow and deep aquifers is mainly controlled by the rock water interaction. In the southwestern part, along with rock-water interactions, the evaporation processes also played an important role (Pant et al., 2020c), as shown in Figure 6. The saturation index discusses about the mineral dissolution and precipitation processes by measuring the equilibrium between minerals and water. Saturation index data showed that a variety of processes (including silicate mineral dissolutions, carbonate mineral weathering, the common ions effect, evaporation, temperature, penetration of wastewater, and irrigation return flows, etc.) supersaturated the groundwater in the Malwa region with the minerals calcite and dolomite (CGWB, 2013). In alkaline conditions, precipitation of calcite governs fluorite dissolution and high fluoride content in the groundwater, as shown in scheme 1 (Kumar and Singh, 2015b; Saxena and Ahmed, 2001)



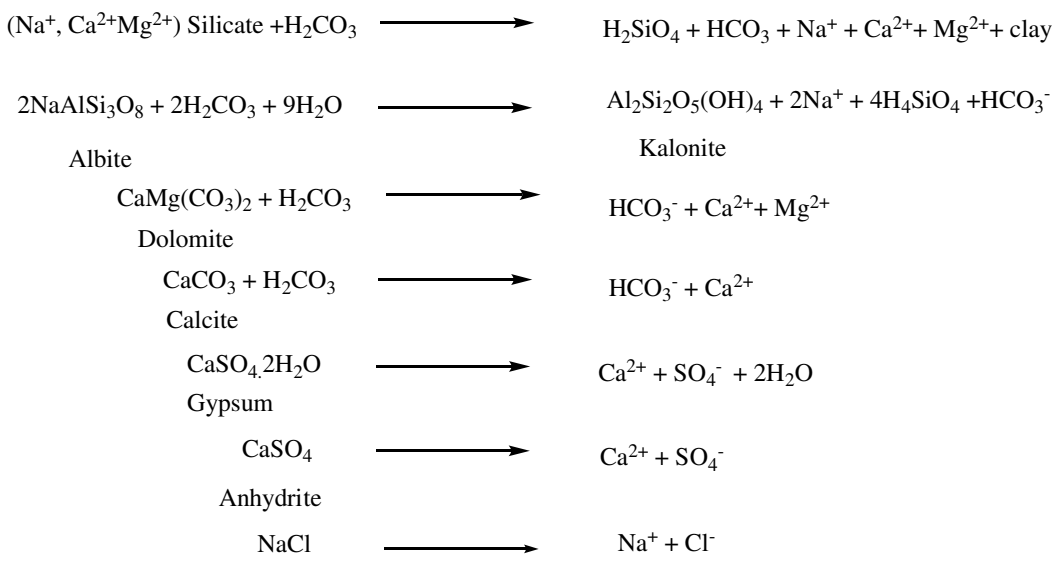
Scheme 1. Calcite precipitation governs fluorite dissolutions

The chemical compositions of groundwater alter during its movement to the subsurface, and these changes are found by the ion exchange index. The ion exchange index describes the exchange of ions either directly or indirectly. In direct ion exchange, Na^+ and K^+ ions from the water exchange with the Ca^{2+} and Mg^{2+} ions from the rock, but in indirect ion exchange, Ca^{2+} and Mg^{2+} ions from the water exchange with Na^+ and K^+ ions from the rock. The groundwater showed reverse ion exchange trends in most Malwa regions: alkali metals such as Na^+ replaced alkaline earth metals (Pant et al., 2020c; Diana Anoubam Sharma et al., 2017). These replacements most commonly happened in clay minerals and can be displayed, as shown in Scheme 2.



Scheme 2. Ion exchange mechanism with clay

The number of direct and indirect interactions between aquifer and bedrock controls the mineral content of groundwater. By software-based bivariate methods, the researcher found that carbonate and silicate minerals' dissolutions control the Malwa region's groundwater chemistry (CGWB, 2020; Singh et al., 2020)



Scheme 3. Silicate, feldspar, dolomite, calcite, fluorite, gypsum and halite weathering.

The silicate weathering occurs upon coming in contact with carbonic acid. The albite ore, silicate weathering and halite dissolutions increased the concentration of sodium ions in groundwater. Dolomite and calcite weathering cause high calcium and magnesium content (Keesari et al., 2014). The gypsum and anhydrite minerals' dissolutions release sulphate ions in water, as shown in Scheme 3. However, there might be other reasons for high sulphate content, such as the breakdown of the organic material and the use of fertilizers (Keesari et al., 2014). In this Indo-Gangetic region of Punjab, sediment deposition occurred due to the erosion of the Himalayan sedimentary rocks by the Indo-Gangetic river system. Each layer contained mixed mineralogic assemblage, and the mineralogic assemblage varied greatly from one region to another (Freeze and Cherry, 1979). The alluvium soil of the Malwa region is made up of sand, silt and clays, and their layering pattern in different areas can be different (CGWB, 2020). Here, a general hypothetical model was designed to understand the various hydrogeochemical reactions operating under the surface, as shown in Figure 7.

As the water moves, it encounters several types of minerals. First, the high oxygen content decays the organic matter in the uppermost layer, and the excess water content releases the

bicarbonate ions in the aquifer (Singh et al., 2020). The groundwater of the Malwa region is hard, and calcium carbonate nodules are there (Masuda et al., 2010). The movement of groundwater through limestone, shown as the 2nd layer in Figure 7, calcite or dolomite dissolution occurs, and water becomes rich with Ca-HCO_3^- type composition. Clay (3rd layer in Figure 7) is rich in quartz, montmorillonite and feldspar-type minerals (Jassal et al., 2001). The interaction of these minerals leads to the supersaturation of calcite ore due to the common ion effect. As the water moves through the gypsum bed (4th layer in Figure 7), sulphate dissolution occurs, and calcite precipitation leads to the re-establishing of calcite equilibrium and sulphates becoming dominant ions (Ahada and Suthar, 2018; Kaur et al., 2017). Bonsor et al. proposed that the saline nature of aquifers was due to the dissolution of halite minerals. Because of the high solubility of these minerals in the water, the groundwater becomes enriched with sodium and chloride ion concentrations (Bonsor et al., 2017).

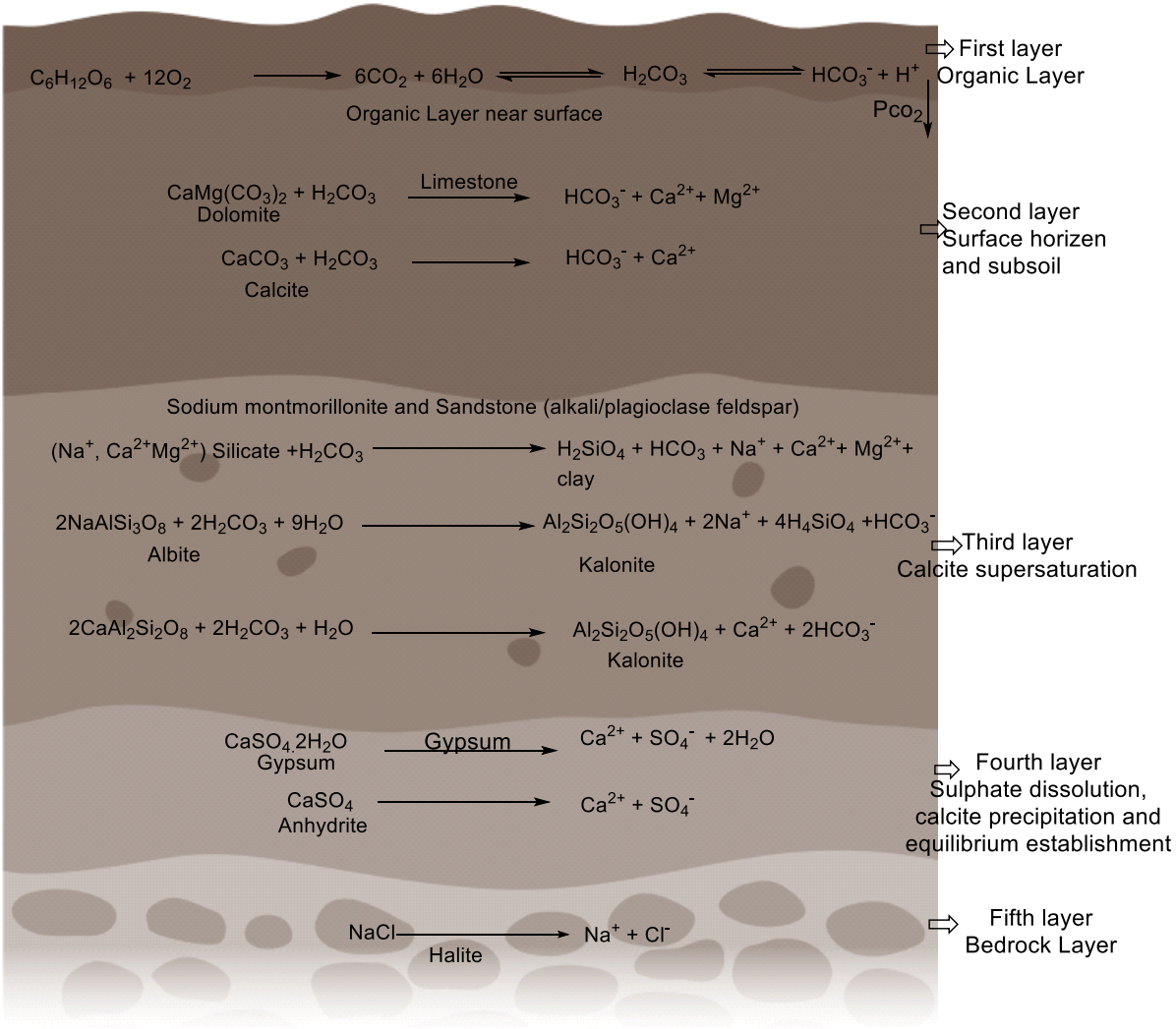


Figure 7. Hypothetical model of various hydro-chemical evolution undergoing the groundwater. The first layer is the uppermost layer, rich in organic matter. The dolomite and calcite-type minerals are denoted in 2nd layer. The clay layer rich in montmorillonite and feldspar-type minerals is demonstrated as the third layer. The dissolutions of gypsum/anhydrite and calcite precipitation are represented as the fourth layer. Finally, the halite-type minerals' dissolutions are embodied as the fifth and last layer.

The chemistry operating under the surface impacts the mobilization of other elements such as U, As, and other heavy metals. After a thorough study, Acosta and co-workers validated that salinity increased the movement of heavy metals in the groundwater (Acosta et al., 2011). Here,

we are limiting our study to two elements, arsenic and uranium and trying to describe their mobilization in groundwater.

Chemical mobilization of arsenic in the aquifer of Malwa regions

In Malwa region aquifers, arsenic is found in neutral arsenite (H_3AsO_3) and arsenate ($H_2AsO_4^-$) forms (CGWB, 2014). In the Himalayan range, arsenic-rich pelitic and argillaceous rocks are commonly found. During the late Pleistocene and early Holocene, weathering processes led to deposit these materials as sediments in Pleistocene alluvium and Holocene alluvium (Herath et al., 2016). The movement of these minerals in the groundwater is mainly controlled by pH, organic matter reduction, redox reactions, and adsorbents such as oxides and hydroxides of iron, manganese, aluminium and clay minerals (Bauer and Blodau, 2006). The As(III) form is more prevalent and harmful in reducing environments. After the green revolution, frequent tube wells were dug for drinking and irrigation water diffusing atmospheric oxygen into this region's aquifers. This diffusion will be resulted in changes in groundwater chemistry and thus causes the oxidation of As(III) into As(V), as shown in Figure 8. (Hundal et al., 2007; Welch and Lico, 1998)

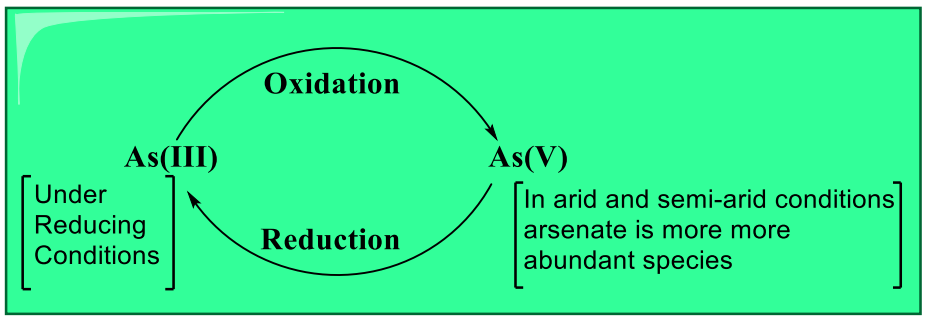


Figure 8. illustration of redox transformation of arsenic in aquifer sediments.

The various geochemical and biological processes played a crucial role in mobilizing and transforming arsenic in the groundwater. In the proposed schematic representation, shown in Figure 9, we tried to show the multiple reasons for releasing arsenic in the groundwater.

Dissolved organic matter is one reason for releasing arsenic from the soil and sediment of aquifers (Sharma et al., 2011). Frequent withdrawal of water from the aquifers by tube-well diffuses oxygen in the aquifer, and oxygen oxidizes the arsenopyrite (FeAsS) and pyrite (FeS_2) minerals, as shown in Figure 5 (reaction 1**,** Figure 9) (Shankar et al., 2014). The Fe(III) deposited in the aquifers can also oxidize these minerals and be the reason for the mobilization of arsenic to groundwater (reaction 2, Figure 9) (Welch and Lico, 1998). According to the report by CGWB, the oxidation of FeAsS and dissolution of Fe(OOH)-As is the most probable reason for arsenic contamination in the groundwater (CGWB, 2014). Nitrate leaching can also oxidize these minerals at low pH (reaction 3, Figure 9) (Zhang et al., 2020). The ferrihydrite sulfidization also liberates arsenites in aquifers (reaction 4, Figure 9) (O'Day et al., 2004). After the reductive dissolution of iron oxides containing orpiment minerals discharges arsenite in the groundwater (reaction 5, Figure 9) (Wang and Mulligan, 2006).

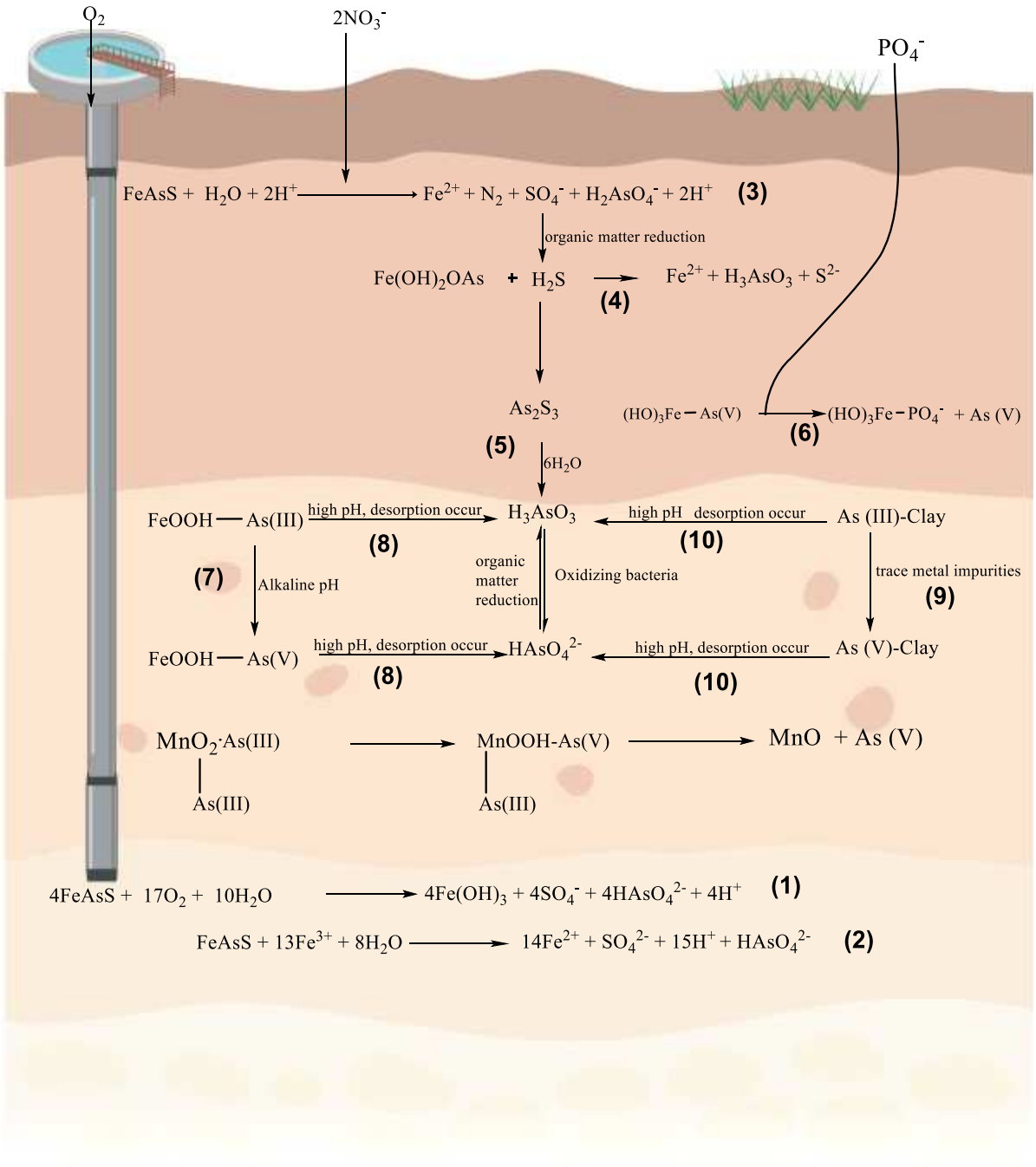


Figure 9. Proposed chemical illustration of various mechanisms for the mobilization of arsenic in groundwater.

The phosphate-containing chemical fertilizers leached through the soil. During the downward movement, phosphate ions react with arsenic-adsorbed minerals to replace arsenic from adsorbed surfaces and let go of the arsenate in the groundwater (reaction 6, Figure 9) (Cui and

Weng, 2013). The $Fe(OH)_3$, $Al(OH)_3$ and clay minerals adsorbed oxyanion of arsenite on their surfaces (Goldberg, 2002; Manning and Goldberg, 1996). The alkaline conditions oxidize the oxyanion species of arsenite to arsenate (reaction 7, Figure 9). At high pH, the desorption of arsenite and arsenate adsorbed on ferric hydroxide's surfaces happened (reaction 8, Figure 9) (Masue et al., 2007). Generally, the arsenate oxyanions are found on clay minerals because trace metal impurities oxidize arsenite to arsenate (reaction 9, Figure 9). Therefore, at high pH, clay-bounded arsenite and arsenate could release into groundwater aquifers (reaction 10, Figure 9) (Frost and Griffin, 1977). Kumar et al. found that in Bathinda, Barnala and Ludhiana, the mean arsenic concentration in groundwater was less after the post-monsoon than pre-monsoon (Kumar et al., 2021), and this might be due to other oxidation-reduction procedures that are described in this schematic diagram. Here, we tried to include all possible ways for the mobilization of arsenic in groundwater. We believe our proposed schematic flowchart will be helpful for the researcher and policymakers in mitigating the arsenic problem.

Chemical mobilization of uranium in the aquifer of the Malwa region

The radiological and toxicological impact of uranium is shattering the socioeconomic model of society (Coyte et al., 2018; Sahoo et al., 2021). To deal with this dangerous metal, a roundtable discussion of experts from different areas, such as chemists, physicists, microbiologists, geologists, zoologists, botanists, and pedologists, is required. But initially, it's essential to understand the chemical mechanism for its mobilisation; thus, an attempt was made. Indeed, the interaction of soil and water minerals with uranium species is changing every day (Ginder-Vogel and Fendorf, 2007). To understand this complicated natural chemical and physical interaction system, we proposed a chemical model of uranium mobilization in the aquifers of the Malwa region, as shown in the schematic diagram (Figure 10). Uranium exists in two forms uranous or uranium^{4+} [U(IV)] and uranyl or uranium^{6+} [U(VI)]. U(VI) is more mobilized and generally found in water, whereas U(IV) is relatively insoluble and makes stable

compounds (Ginder-Vogel and Fendorf, 2007; Qafoku and Icenhower, 2008). $UO_2(CO_3)_3^{4-}$ and $(UO_2)_3(OH)_5^+$ are the two forms that are frequently found in the groundwater of the Malwa region. The mobilization of these uranium forms in groundwater depends upon various chemical and physical factors such as the climate of that region, rock-water interaction, hydrogeochemical conditions (Langmuir, 1978). It isn't easy to single out one particular reason responsible for these forms in the groundwater. So, we tried to introduce all possible sources for uranium dissolution in the Malwa region's groundwater. In the presence of oxygen and highly alkaline conditions, U(VI) is more prevalent and exists as $UO_2(CO_3)_3^{4-}$ and at pH above 8.5, the latter species exists in equilibrium with $(UO_2)_3(OH)_5^+$ (reaction 2, Figure 10) (Langmuir, 1978). In the presence of high calcium ion concentration, uranyl carbonyl ion form $Ca(UO_2)(CO_3)_3^{2-}$ (reaction 4, Figure 10) and this can also form from $Ca_2(UO_2)(CO_3)_2$ in the presence of bicarbonate ion concentration and oxic conditions (reaction 5, Figure 10) (Dong and Brooks, 2006). The oxidation of liebigite ore also yields the same result (reaction 6, Figure 10) (Gorman-Lewis et al., 2008). At neutral conditions (pH = 7) and high phosphate ion concentrations, the equilibrium between Uranophane and Autunite shifts towards autunite (Cuney, 2010; Langmuir, 1997). Autunite is one of the predominant species around pH 6-7.5 (reaction 8, Figure 10) (Langmuir, 1997). However, uranyl carbonyl complexes predominate at higher pH and high carbonate ion concentrations (reaction 9, Figure 10) (Barnett et al., 2000; Phillippi et al., 2007). Through the process of biosorption, uranium ion sorption occurs on the organic matter and forms UO_2(organic matter) complexes (Newsome et al., 2014; Tsezos and Volesky, 1982). Still, in the presence of a sufficient amount of carbonates/bicarbonates ions, this UO_2(organic matter) dissolution happens, and uranyl carbonyl complexes form (reaction 1, Figure 10) (Cumberland et al., 2016).

The insoluble uraninite converts into uranyl carbonyl complexes in the presence of carbonate/bicarbonate ions, iron oxides (FeOOH, Fe_2O_3), and an oxic environment (reaction 3,

Figure 10) (Stewart et al., 2015). The oxidation of uraninite ore leads to the formation of uranyl ionic species (reaction 10, Figure 10) (Bala et al., 2022). The latter ions in high carbonate concentrations form uranyl carbonate complexes that mobilize in the groundwater (reaction 26, Figure 10) (Chandrasekar et al., 2021). Introducing oxygen and nitrate oxidizes the reduced Fe^{2+} to amorphous FeOOH, which can oxidise the uraninite (Senko et al., 2005).

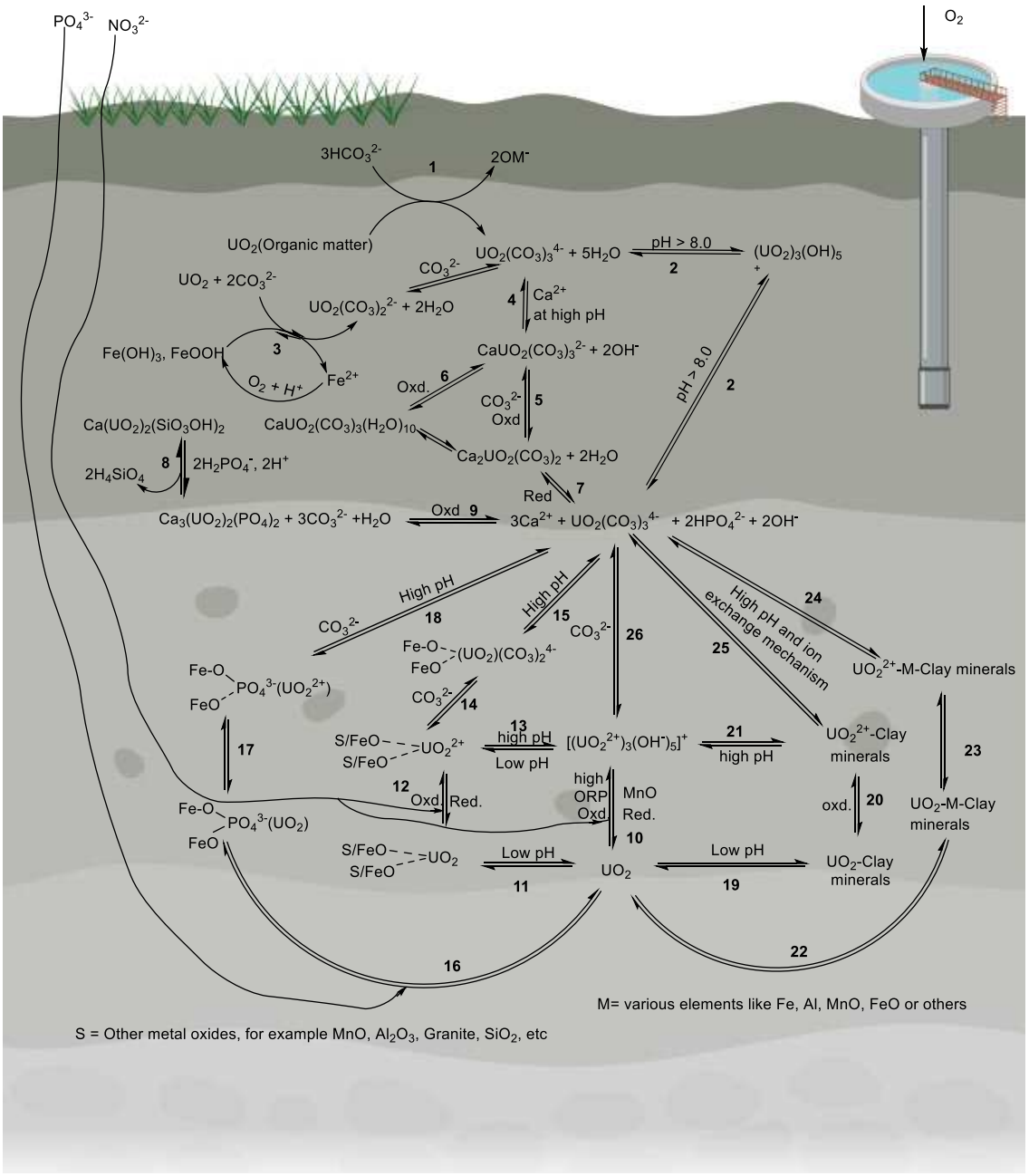


Figure 10. Proposed chemical illustration of various mechanisms for the mobilization of uranium in groundwater.

All over the globe, the deposits of uranium ores were identified within the granite plutons, roll-front deposits, sandstones, breccia and organic matters ((Cumberland et al., 2016) and reference cited therein). In these deposits, the uranium exists as insoluble uraninite form UO_2 adsorbed on the surfaces of minerals like iron oxides (FeO), manganese oxides, alumina, gibbsite, granite, quartz and natural sediments etc [(Qafoku and Icenhower, 2008) and references cited therein]. The literature data showed that uranium adsorbed on the surfaces of FeO as bidentate or tridentate complexes at low pH, but bidentate complexes are more common (reaction 11, Figure 10) (Ching-kuo Daniel Hsi and Langmuir, 1985). The adsorbed uraninite on the surface of FeO oxidized by NO_3^-, microbial oxidation, O_2 or other factors changes into uranyl [U(VI)] complexes (reaction 12, Figure 10) (Bonotto et al., 2019; Liesch et al., 2015). At high pH, desorption of uranyl ions occurs, and uranyl ions mobilize in the groundwater (reaction 13, Figure 10) (Ching-kuo Daniel Hsi and Langmuir, 1985). The surface-adsorbed uranium complexes sometimes also make complexes with carbonate ions (reaction 14, Figure 10), but these complexes are not stable (Bargar et al., 2000). Above pH 8, the adsorption affinity of these complexes towards the FeO surfaces decreases, and uranyl carbonate complexes dissolve into the water (reaction 15, Figure 10) (Wazne et al., 2003). Similar studies reported other metal oxides, for instance, MnO (Wang et al., 2013), Al_2O_3 (Sylwester et al., 2000), Granite (Baik et al., 2004), SiO_2 (Reich et al., 1998), etc. Sometimes bidentate FeO adsorbs PO_4^{3-} ions and makes $(FeO)_2PO_4^{3-}$ type complexes that show better affinity and holding capacity for uranium ions than simple FeO (reaction 16, Figure 10) (Del Nero et al., 2011; Finch and Murakami, 1999; Seder-Colomina et al., 2015). However, in oxic conditions, this bounded uraninite form converts into FeO-associated uranyl [U(VI)] phosphate complexes (reaction 17, Figure 10). According to literature data, PO_4^{3-} has 3 to 4 orders of lower affinity

than carbonates (Sahoo et al., 2022). Hence in the presence of high carbonate ion concentration and high pH, these uranium species dissolve as uranyl carbonyl complexes (Langmuir, 1978; Wazne et al., 2003).

Sediments and soil are both repleted with clay minerals. These clay minerals showed strong chemical and physical interactions with the dissolved species because of small particle size, complex porous structure, high specific surface area (Schulze, 2018). Uraninite is also adsorbed on the clay minerals at low pH (reaction 19, Figure 10) (Davey and Scott, 1956; Hennig et al., 2020). However, U(IV) is readily converted into a uranyl-clay complex by microbial oxidation or other oxidation processes (reaction 20, Figure 10). This uranyl clay complex mobilized in the water as $([(UO_2^{2+})_3(OH^-)_5]^+$ at high pH (reaction 21, Figure 10) (Bachmaf and Merkel, 2011). In addition, clay minerals have a high negative charge on their surface, so they adsorb positive metal ions or metal oxides (Geckeis et al., 2013). These metal or metal oxide ions provide better space for holding uranium species (reactions 22 and 23, Figure 10) (Catalano and Brown, 2005; Křepelová et al., 2007; Payne et al., 2004). But at high pH conditions, by ion exchange mechanisms, these oxides liberate uranium in the groundwater as uranyl carbonate complexes (reactions 24 and 25, Figure 10) (Greathouse and Cygan, 2005; Křepelová et al., 2006). In the three districts of the Malwa region, Kumar et al. compared the uranium ion concentration in the pre-monsoon and post-monsoon periods. They found that the mean uranium concentration decreased after the post-monsoon than pre-monsoon; this might be due to the various oxidation and reduction reactions described in Figure 10.

Technological developments for mitigating these issues

The SciFinder database search found over 150 thousand articles about contaminations, toxicity and mitigation/removal methods of arsenic and uranium. Nonetheless, the situation of the millions of individuals affected by uranium and arsenic remains unchanged. According to a

world bank report, low-middle-income countries like India suffered the most from these problems (World Bank Group, 2020). As shown in the upper described schemes, the dissolution of arsenic and uranium in the groundwater is more related to the oxidation and reduction processes (Jain and Singh, 2012). Hence, in the past and recent research databases, we found a range of technologies (from conventional to advanced) available to treat arsenic/uranium-rich groundwater (Gandhi et al., 2022; Yadav et al., 2022). These technologies are based on various processes, such as ion exchange, membrane filtration, electrocoagulation, photocatalysis, adsorptions, co-precipitation and biological methods (Dinis and Fiúza, 2021). Many excellent reviews have been published elsewhere describing these processes and technologies (Hao et al., 2018; Jain and Singh, 2012; Katsoyiannis and Zouboulis, 2013; Weerasundara et al., 2021; You et al., 2021).

Over the last few years, there has been tremendous technological advancement in uranium and arsenic removal processes. But it is necessary to consider a location-specific uranium and arsenic mitigation strategy. Due to variances in geography, geomorphology, and socioeconomic and literacy levels of the populace, a solution that works in one location should not be universally applied to the other afflicted places. Even the laboratory protocols showed limitations, like the protocols/procedure developed in one laboratory yielded different outcomes in another laboratory and field. Many technologies demonstrated encouraging results in pilot studies but are ineffective in real-world settings.

The high cost of adsorbents and interference due to competitive ions for adsorption made the adsorptions methods costly and economically unviable. Although the membrane filtrations and ion-exchange methods are effective in uranium and arsenic removal, the high initial cost and recurring cost made these methods unfeasible for low-middle-income countries. The generation of sludge is a big problem with the chemical coagulation with metal salts and lime followed by filtering, even though it is a highly effective, affordable, and popular procedure.

Biosorption and bioremediation showed quite exciting results at the laboratory scale. With low uranium concentrations, electrocoagulation and photocatalysis were also effective techniques. The choice of a process is further considerably influenced by factors such as process complexity, management of hazardous materials, and waste disposal. Waste generation and disposal are significant concerns with these technologies. In table 5, we tried to explain the various technology available with their merit, demerits, efficiency and running cost.

Table 5. Analysis of various technologies for the arsenic and uranium remediation process.

S. N	**Treatment Process**	**Metal**	**Methods for uranium and arsenic removal**	**Advantage**	**Disadvantage**	**E*%**	**RC*(US$/m^3)**	**Ref**
1	Adsorption	Ar	Iron-based sorbents, activated carbon, clays and soils, aluminium-based sorbents, zeolites and other various adsorbents	Extensively used in treatment because of its low initial cost, ease of operation and low power requirements.	Other contaminants lower its productivity. Adsobents cost is also an important factor.	60-99	0.10-1.19	(Awual et al., 2019; Hao et al., 2018; Lata and Samadder, 2016; Weerasundara et al., 2021)
		U	Amine functionalised Clays, modified metal-organic frameworks, modified graphene oxides, silica-coated nanoparticles			77-100		(Tobilko et al., 2019)(Gandhi et al., 2022 and references cited therein)
2	Ion exchange	Ar	Using different Ion exchange materials or membranes example, TiO_2-loaded Amberlite resin etc.	Highly efficient methods and commercially available	Not suitable for high TDS water. Quite costly material of membranes and membrane needs removal after saturation. Harmful waste generation	95%	0.12 as per laboratory conditions	(Awual et al., 2013, 2008; Awual and Jyo, 2009)
		U	Zirconium phosphate resins, chelax-100, Dowex resin, ORWA resin, synthetic resins etc.			94-99.5		(Gandhi et al., 2022)(Florez et al., 2017)
3	Membrane filtration	Ar	Four types of membrane processes: microfiltration, ultrafiltration, nanofiltration and reverse osmosis	Best and highly efficient technology available	Very ionic strength reduces its capacity, requires skilled workers, and the need for pH adjustment and the addition of PH control equipment increases the overall cost of the process. Solid waste regeneration		0.52-0.88	(Algieri et al., 2022) (Richards et al., 2022)
		U				90-99		(Dinis and Fiúza, 2021)

4	Chemical Coagulation and chemical reduction	Ar	Titanium chloride, aluminium, iron, and zirconium chloride-based coagulants.	Readily available material, simple process, and does not require a trained individual	Produce large amounts of arsenic and uranium-bearing sludge, hence need proper disposal. Sometimes pH treatment is required. The overall cost adds up.	Upto 90	0.076 without sludge disposal	(Awual et al., 2012, 2011) (Ahmad et al., 2018; Cañas Kurz et al., 2020)
		U	Ferric sulphate, ferrous sulphate, aluminium sulphate, barium chloride, lime softening, iron chloride etc.			68-99		(Dinis and Fiúza, 2021)(Katsoyiannis and Zouboulis, 2013)
5	Electro coagulation	Ar	Iron Electrodes	99-99.65% removal possible	Generated hazardous sludge needs proper disposal	99-99.65	0.22-0.31	(Kabir and Chowdhury, 2017)
		U	Magnesium, aluminium and iron electrodes in the presence of organic ligands					(Carolin et al., 2017; Choi et al., 2020)
6	Biological Remediation	Ar	Removal by phytoremediation, bacteria, microalgae and fungi	Cost-effective and no need to remove generated biomass Phytoremediation is an efficient, environmentally friendly and low-cost technique.	A large volume of culture is required. It is a tedious process. Treated water needs tertiary treatment that enhances its overall cost. For phytoremediation, skilled workers/ subject experts are needed. Moreover, a large area and more treatment time required		-	(Ghosh (Nath) et al., 2019)
		U				30-85.8		(You et al., 2021) (Dushenkov et al., 1997)
7	Hybrid methods	Ar	First oxidation by biological means followed by adsorption etc.	Highly efficient technique	Absorbents used in this process cost more than 50% of the total operational cost.	100 %	0.15-0.19	(Katsoyiannis et al., 2015)
8	Photocatalysis	U	by irradiating light onto the semiconductor surface U(VI) reduced into U(IV)	100% removal possible	The high cost and need for specific treatment make this uneconomical but advances going on in this area	77-100 %	-	(J. Wang et al., 2020; Z. Wang et al., 2020)

Conclusions and recommendations

Overall, changes in physical and chemical conditions cause the change in under-surface chemistry operating in the aquifer of any region. Here, we looked at the Malwa region of Punjab, India, where overexploitation and anthropogenic activity modified the chemistry beneath the surface and caused dangerous substances like uranium and arsenic to mobilize in

the water. Through this paper, we tried to explain the various chemical changes below the earth's surface by a hypothetical model and how these changes affect the mobilization of arsenic and uranium in the groundwater of the Malwa region. To mitigate these problems, the central and state governments made several policies and laws to handle the water pollution problems in Punjab. The various government departments and agencies such as the Indian Council of Medical Research (ICMR), Punjab Pollution Control Board (PPCB), Central Groundwater Board (CGWB), Department of Drinking Water Supply and Sanitation (DWSS), Bhabha Atomic Research Centre (BARC), Department of Agriculture (DoA), Economic Protection Agency (EPA), Economic and Statistical Organization of Punjab (ESOP) are conducting their research and take appropriate actions. In their efforts, we believe chem-environmental views of our designed models will help better understand the issues and take the appropriate action.

For the mitigation of these issues, numerous technological advancements have been made. However, their installation and recurring maintenance cost made these technologies unaffordable and unsustainable for long-term use, particularly in low- and middle-income nations (Yadav et al., 2022), especially in the Malwa region, where family income is overburdened due to disease-like cancer. It's difficult to purchase and use these technologies. Hernandez et al. suggested that the most efficient solutions might be achieved by implementing community-based removal plants with the help of effective policy-level initiatives (Hernandez et al., 2019). Long-term objectives of the policy-level intervention could include raising community awareness and promoting greater access to hygienic facilities and clean water for all. So, to address the issue on a greater scale, coordinated policy-level initiatives with community participation are required alongside continued research on developing improved and better technologies for removal methods. Moreover, Governments and their agencies should implement the proper control measures, such as

- Based on the above information about groundwater pollutants, the DWSS may strengthen monitoring systems to ensure a prompt and effective response, be more proactive in controlling sources in affected areas to provide safe drinking water, and effectively engage local populations to ensure that clean water is used.
- Provide a clean piped water supply for drinking and cooking and ensure water is stored and handled correctly.
- The Gram Panchayat Water and Sanitation Committees (GPWSCs) should be aware of the problems posed by poor water quality and actively participate in managing the water supply systems, especially ensuring proper disinfection. Create awareness at the individual, community and institutional levels.
- After all other groundwater and surface water options have been tried, treatment technologies should only be used as a last resort.

Finally, the need of the time is for better interdepartmental strategy and inter-disciplinary approach and research with sound science and adaptive policy and management practice to mitigate uranium and arsenic problems throughout the globe. Our designed models will help policymakers and academicians/scientists take proper control measures and develop more and better technologies in the future to tackle these problems globally.

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Author contribution

Vajinder Kumar conceived the idea and wrote the whole manuscript. Arnab Maity, Avneesh Kumar, Sandip Saha, and Paul Kay helped in writing the manuscript through discussions and idea exchange, and value addition. Baljinder Singh and Tirtha Mukherjee read the manuscript and gave valuable comments on the manuscript.

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